

Quantum Phase Transition in the Itinerant Antiferromagnet $(V_{0.9}Ti_{0.1})_2O_3$

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(Received 9 May 2008; published 29 August 2008)

Quantum-critical behavior of the itinerant electron antiferromagnet $(V_{0.9}Ti_{0.1})_2O_3$ has been studied by single-crystal neutron scattering. By directly observing antiferromagnetic spin fluctuations in the paramagnetic phase, we have shown that the characteristic energy depends on temperature as $c_1 + c_2 T^{3/2}$, where c_1 and c_2 are constants. This $T^{3/2}$ dependence demonstrates that the present strongly correlated d -electron antiferromagnet clearly shows the criticality of the spin-density-wave quantum phase transition in three space dimensions.

DOI: [10.1103/PhysRevLett.101.096406](https://doi.org/10.1103/PhysRevLett.101.096406)

PACS numbers: 71.27.+a, 71.10.Hf, 75.40.Gb

In recent years, novel viewpoints of matter have been exploited by quantum phase transitions (QPT) [1,2], zero-temperature second-order phase transitions tuned by pressure or other controlling parameters. Around a QPT, the state of matter is characterized by singular behavior of fluctuating order parameters having both quantum mechanical and thermal origins. Quantum phase transitions are investigated in broad fields ranging from high temperature superconductors [3,4], metal-insulator transitions [5], to heavy fermions [6,7]. Although a number of QPTs have been investigated experimentally and theoretically, many problems are under controversial debates.

A QPT separating a ferromagnetic (FM) or antiferromagnetic (AFM) state to a paramagnetic state in an itinerant electron system has been studied for decades. Its theory was first developed by Moriya and coworkers [8–10]. The modern formulation of this theory using renormalization group techniques was provided by Hertz [11,12]. The theoretical predictions of the FM QPT are in general supported by the experimental studies of, for instance, d -electron FM metals MnSi and ZrZn₂ [8,13,14]. However, recent studies of the FM QPT have shown that there are important perturbative effects closer to the critical point [7,15,16].

For the itinerant AFM QPT, referred to as the spin density wave (SDW) QPT, the problem is more complicated and is not settled. Experimentally, thermodynamic and transport properties studied on, e.g., d -electron AFM metals β -Mn, V₃Se₄ [8], and f -electron AFM heavy fermions [6,7] are in rough agreement with theories of the SDW QPT. However, most neutron scattering studies seem to contradict expectations of the SDW QPT [1]. For example, observed AFM spin fluctuations of the heavy fermion CeCu_{6-x}Au_x [17] exhibit E/T scaling, suggesting the existence of a new type of QPT [1,7,18,19]. On the other hand, our recent neutron scattering study on the

heavy fermion Ce(Ru_{1-x}Rh_x)₂Si₂ is consistent with the SDW QPT with no indication of E/T scaling [20]. Therefore, there are many open questions on QPTs for itinerant antiferromagnets, such as, whether the SDW QPT can be applicable to the itinerant d - and f -electron AFM systems, or how fundamentally new QPTs are formulated to account for the complexity of experimental data of these itinerant systems [6,7,16,19,21,22].

The isomorphous weak AFM metals V_{2-y}O₃ [23] and (V_{1-x}Ti_x)₂O₃ [24] belong to the celebrated Mott-Hubbard system (V_{1-x}M_x)₂O₃ ($M = \text{Cr, Ti}$) [25], which shows metal-insulator transitions due to strong correlation effects (Fig. 1) [5]. The $3d^2$ electronic state of the V³⁺ ion is in an $S = 1$ high spin state with an effective moment $\sim 2.8\mu_B$ [26,27]. For the AFM metallic (V_{1-x}Ti_x)₂O₃ ($x > 0.05$), only a small fraction of the moment $\sim 0.3\mu_B$ forms the AFM ordering below $T_N = 23$ K ($x = 0.1$) [24]. The second-order AFM transition is tuned to a QPT by hydrostatic pressure of the order of 2 GPa [23,28], and quantum-critical behavior can be expected to be observed in the paramagnetic metallic phase.

Previous neutron-scattering experiments on V_{2-y}O₃ clarified several interesting aspects of this system [23]. At the same time, their results raised some controversy [1]. In those experiments, AFM spin fluctuations were roughly consistent with a SDW QPT, while the data suggested the E/T scaling indicating a novel QPT. However, the statistical accuracy of those experiments was not sufficient for drawing a definite conclusion on the QPT. Thus in this work, we reinvestigate the AFM quantum-critical behavior in the paramagnetic metallic phase using (V_{0.9}Ti_{0.1})₂O₃ [24], which is suited for the present purpose because its local disorder is weaker than in V_{2-y}O₃. By sufficiently improving the statistical accuracy, we have concluded that the AFM spin fluctuations agree well with those of the SDW QPT in three space dimensions.

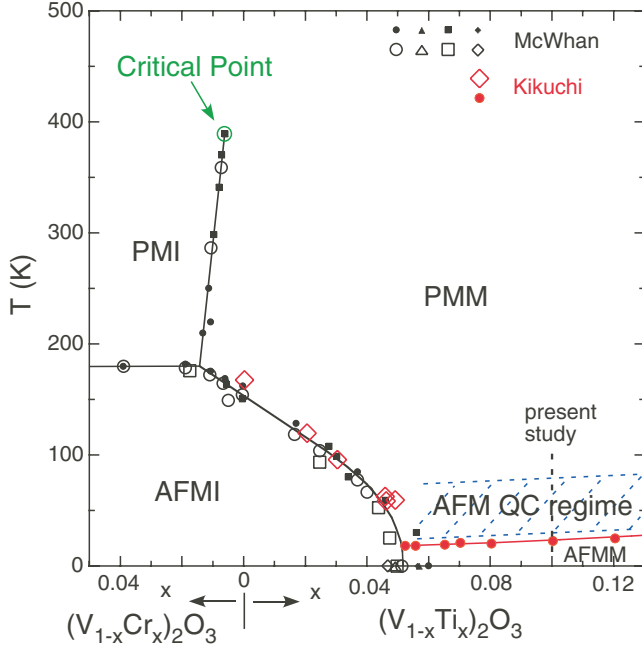


FIG. 1 (color online). Phase diagram of $(V_{1-x}M_x)_2O_3$ ($M = \text{Cr, Ti}$) is reproduced using data points of Refs. [24,25]. PMI, PMM, AFMI, and AFMM stand for paramagnetic insulator, paramagnetic metal, antiferromagnetic insulator, and antiferromagnetic metal phases, respectively. AFM QC regime, inferred from the present study for $x = 0.1$, is the temperature range where the quantum-critical AFM fluctuations are controlled by the SDW QPT. The AFM transition between PMM and AFMM is a second-order phase transition which is tuned to the QPT by hydrostatic pressure [23,28].

Neutron-scattering measurements were performed on the triple-axis spectrometers ISSP-GPTAS at the Japan Atomic Energy Agency, BT-7 at the NIST Center for Neutron Research, and HB1 at Oak Ridge National Laboratory (ORNL). They were operated using a final energy of $E_f = 14$ meV, providing an energy resolution of 1.4 meV (full width at half maximum) at elastic positions. A single-crystal sample of $(V_{0.9}\text{Ti}_{0.1})_2O_3$ with a weight of 2 g was grown by the floating zone method. The crystal was mounted in closed-cycle He-gas refrigerators so as to measure a $(H, 0, L) = Ha^* + Lc^*$ scattering plane, where a^* and c^* are the hexagonal reciprocal lattice vectors. All the data shown are converted to the dynamical susceptibility and corrected for the magnetic form factor.

The AFM fluctuations of $(V_{0.9}\text{Ti}_{0.1})_2O_3$ expressed as the imaginary part of the dynamical susceptibility at wave vector $\mathbf{Q} + \mathbf{q}$, where $\mathbf{Q} = (1.90 \pm 0.01)c^*$ is the AFM modulation wave vector [24], are described by the Lorentzian function [23]

$$\text{Im } \chi(\mathbf{Q} + \mathbf{q}, E) = \frac{\chi(\mathbf{Q})\Gamma(\mathbf{Q})E}{E^2 + [\Gamma(\mathbf{Q}) + D(q_c^2 + Fq_{ab}^2)]^2}, \quad (1)$$

where E represents the excitation energy, q_c and q_{ab} are components of \mathbf{q} along the c axis and in the ab plane, respectively, D and F are T independent parameters, $\chi(\mathbf{Q})$

and $\Gamma(\mathbf{Q})$ stand for the wave-vector-dependent magnetic susceptibility and characteristic energy, respectively. This form agrees with the approximation used in the theory [1,3,7,8] of the SDW QPT for small \mathbf{q} and E , provided that the product $\chi(\mathbf{Q})\Gamma(\mathbf{Q})$ is T independent. We note that $\Gamma(\mathbf{Q})$ vanishes at a QPT. In Fig. 2(a), the dynamical susceptibility Eq. (1) is illustrated using parameters at $T = 30$ K, where $\Gamma(\mathbf{Q}) = 0.95$ meV. To confirm this Lorentzian function for $(V_{0.9}\text{Ti}_{0.1})_2O_3$, we carried out constant- E scans along the $\mathbf{q} = (\Delta H, 0, 0)$ and $(0, 0, \Delta L)$ lines at three typical temperatures $T = 30, 50$, and 75 K.

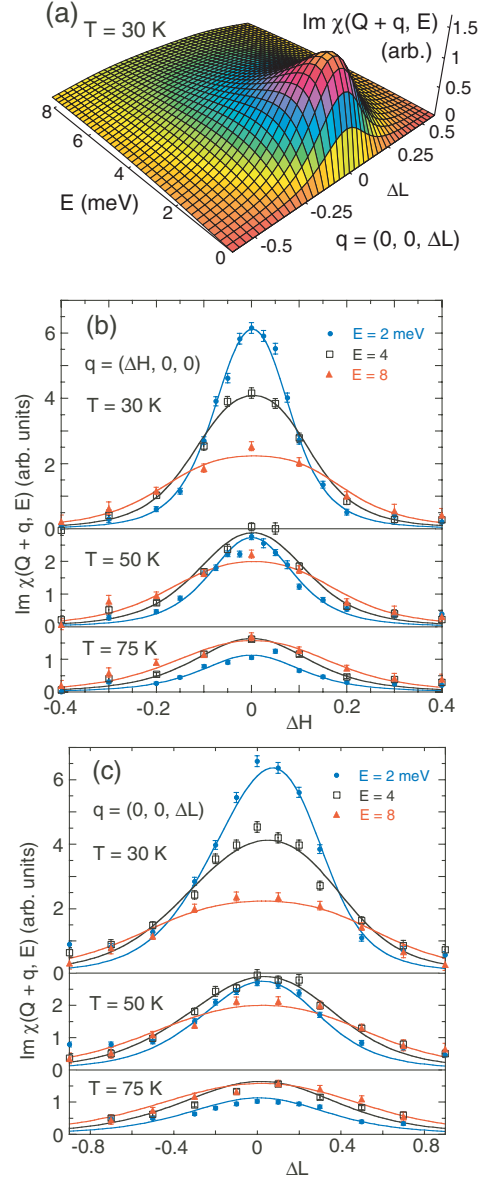


FIG. 2 (color online). (a) Illustration of the quantum-critical behavior of the dynamical susceptibility $\text{Im } \chi(\mathbf{Q} + \mathbf{q}, E)$, Eq. (1), at $T = 30$ K. (b), (c) Constant- E scans taken with $E = 2, 4$, and 8 meV along (b) $\mathbf{q} = (\Delta H, 0, 0)$ and (c) $(0, 0, \Delta L)$ lines at $T = 30, 50$ and 75 K. Curves are fits using Eq. (1) convoluted with the resolution function.

By least squares fitting, we obtained $D = 96 \pm 4$ meV \AA^2 and $F = 0.77 \pm 0.03$. In Figs. 2(b) and 2(c), we show these spectra together with the fit curves of Eq. (1) convoluted with the resolution function. One can see from this figure that Eq. (1) well reproduces the experimental data, in particular, for small q and E . By this reproduction, we confirmed another assumption of Eq. (1) that $\chi(\mathbf{Q} + \mathbf{q})\Gamma(\mathbf{Q} + \mathbf{q})$ does not depend on q .

The theory of the SDW QPT in three dimensions predicts [1,7] that the characteristic energy $\Gamma(\mathbf{Q})$ depends on T as

$$\Gamma(\mathbf{Q}) = c_1 + c_2 T^{3/2}, \quad (2)$$

where $c_1 (<0)$ and c_2 are constants, in the quantum-critical regime $T_N < T \ll T_{\text{coh}}$, where the coherence temperature $T_{\text{coh}} \sim 450$ K [29] represents the effective Fermi energy. It should be noted that the T dependence of $T^{3/2}$ [8,10] in Eq. (2) is the most important characteristic of the SDW QPT. We also note that Eq. (2) breaks down near T_N because the theory neglects the criticality of the finite-temperature phase transition. In an alternative formalism using the self-consistent renormalization (SCR) theory of spin fluctuations [3,8], equivalent to the SDW QPT, the T dependence of $\Gamma(\mathbf{Q})$ is determined by the self-consistent equation

$$\Gamma(\mathbf{Q}) = c'_1 + F_Q \int_0^\infty dE \frac{1}{e^{E/k_B T} - 1} \sum_q \text{Im} \chi(\mathbf{Q} + \mathbf{q}, E), \quad (3)$$

where $c'_1 (<0)$ is a constant and F_Q is the mode-mode coupling constant. This equation employed with Eq. (1) and $\chi(\mathbf{Q})\Gamma(\mathbf{Q}) = \text{const}$ can be used as an experimental fit formula, where c'_1 and F_Q are treated as adjustable parameters.

In order to accurately measure the T dependence of $\Gamma(\mathbf{Q})$, we performed constant- Q scans at the AFM wave vector using better counting statistics than Ref. [23]. The observed spectra were fit to Eq. (1) convoluted with the resolution function. Several spectra and fit curves are shown in Fig. 3, demonstrating excellent agreement between the observation and calculation. Figure 4 shows the T dependence of $\Gamma(\mathbf{Q})$ and $\chi(\mathbf{Q})\Gamma(\mathbf{Q})$ as a function of $T^{3/2}$ and T , respectively. The predictions of the SDW QPT, Eq. (2) and $\chi(\mathbf{Q})\Gamma(\mathbf{Q}) = \text{const}$, which are also plotted using lines in the figure, are in good agreement with the experimental data in the range $1.1T_N < T < 80$ K. By least squares fitting, we obtained $c_1 = -0.37 \pm 0.05$ meV and $c_2 = 0.0083 \pm 0.0002$ K $^{-3/2}$. We also performed the SCR fit using Eq. (3), where $c'_1 = -1.1 \pm 0.2$ meV provided the best fit. This fit curve shown in Fig. 4 also well reproduces the experimental data in the same temperature range. Therefore, we conclude that the AFM spin fluctuations of $(\text{V}_{0.9}\text{Ti}_{0.1})_2\text{O}_3$ in $1.1T_N < T < 80$ K, which can be regarded as the quantum-critical regime, are well ac-

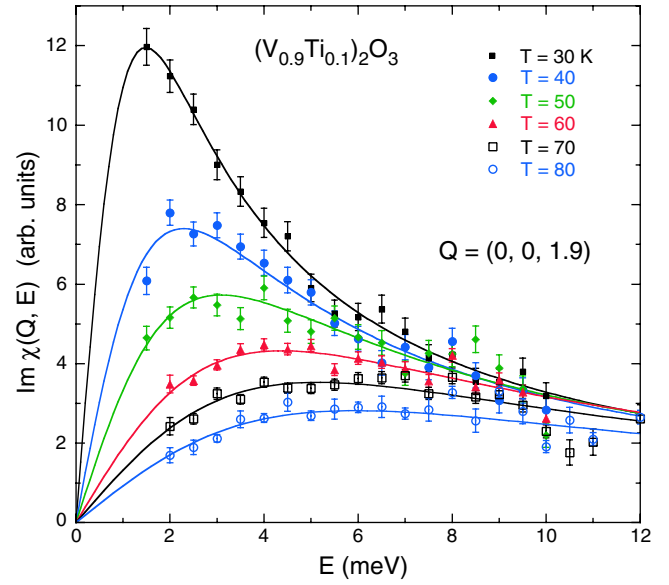


FIG. 3 (color online). Constant- Q scans measured at the AFM wave vector $\mathbf{Q} = 1.9\mathbf{c}^*$ at several temperatures. Curves are fit results using Eq. (1) with two adjustable parameters $\Gamma(\mathbf{Q})$ and $\chi(\mathbf{Q})$. Error bars are statistical in origin and represent 1 standard deviation.

counted for by the quantum-critical behavior of the SDW QPT in three dimensions.

It should be noted that the theories of SDW QPTs are based upon the single-band Hubbard model in a weak correlation regime [3,8,10–12]. However, the electronic state of V_2O_3 is represented by a three-band model with

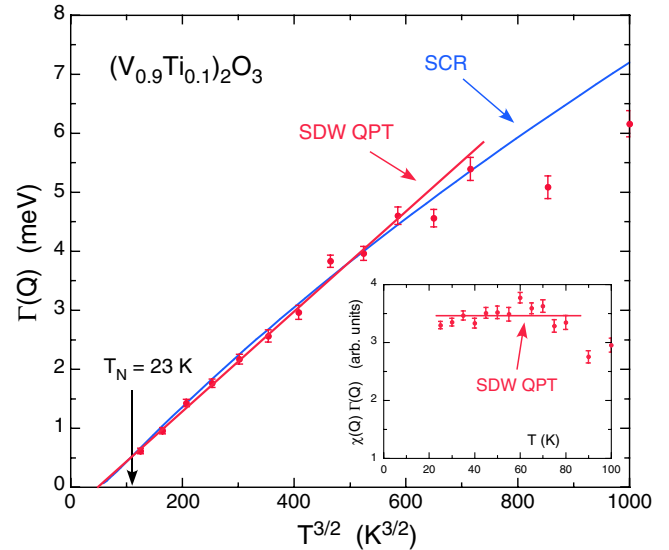


FIG. 4 (color online). Temperature dependence of the characteristic energy $\Gamma(\mathbf{Q})$ of the AFM spin fluctuations is plotted as a function of $T^{3/2}$. The curves represent the prediction Eq. (2) for the SDW QPT and the fit using the SCR theory Eq. (3). The inset shows temperature dependence of the product $\chi(\mathbf{Q})\Gamma(\mathbf{Q})$. The straight line $\chi(\mathbf{Q})\Gamma(\mathbf{Q}) = \text{const}$ is the prediction of the SDW QPT.

strong correlation [26]. The two $3d$ electrons in the V^{3+} ion occupying three degenerate t_{2g} orbitals are coupled by a strong Hund's rule exchange interaction, which gives rise to the $S = 1$ state and the orbital degrees of freedom [26]. The prominent quasiparticle peak at the Fermi energy observed by photoemission spectroscopy [27] and the low coherence temperature $T_{\text{coh}} \sim 450$ K [29] underline the importance of the strong correlation in V_2O_3 . Thus, the present result poses a natural question whether the paramagnetic metallic state of the realistic three-band model shows the same quantum criticality as the SDW QPT. We note that T_{coh} is comparable to the temperature scale $T_0 \sim 320$ K of the SCR theory [8,30], and that the upper bound temperature 80 K of the quantum-critical regime in $(V_{0.9}Ti_{0.1})_2O_3$ may be partly ascribed to orbital fluctuations [23], which are neglected in the theories of the SDW QPT.

It is widely accepted that the correct understanding of AFM QPTs is essential for studying unconventional superconductivity which has been found in an increasing number of strongly correlated electron systems, including high- T_c cuprates, heavy-fermion, and organic superconductors, e.g., $La_{2-x}Sr_xCuO_4$ [31]. In these systems, attractive electron couplings were proposed to be ascribed to AFM spin fluctuations [3,19]. In this context, the spin fluctuations observed in $(V_{0.9}Ti_{0.1})_2O_3$ can be considered as a simple nonsuperconducting case [30].

In conclusion, neutron scattering shows that the quantum-critical spin fluctuations in the paramagnetic metallic phase of the Mott-Hubbard system $(V_{0.9}Ti_{0.1})_2O_3$ agree well with the theoretical predictions of the SDW QPT in three dimensions. The present work is the first clear verification of the SDW QPT in a d -electron itinerant antiferromagnet. The present finding and our recent similar result of an f -electron heavy fermion [20] imply that a broader theoretical basis for the SDW QPT is required to include multiband models and strong correlation effects. Further investigations of the AFM long-range ordered state close to the SDW QPT and crossover phenomena of the QPT to finite-temperature phase transitions will be interesting.

We acknowledge discussions with T. Moriya and Y. Tabata. Work on BT7 and HB1 was supported by the US-Japan Cooperative Program on Neutron Scattering. The authors are grateful for the local support staff at NIST and ORNL. The work at ORNL's High Flux Isotope Reactor was sponsored by the Scientific User Facilities Division, office of Basic Energy Sciences, US Department of Energy.

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